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Accuracy of AlGaAs growth rates and composition determination using RHEED oscillations ☆

T.E. Harvey[a,*], K.A. Bertness[a], R.K. Hickernell[a], C.M. Wang[b], J.D. Splett[b]

[a] *Optoelectronics Division, National Institute of Standards and Technology,815.04, 325 Broadway, Boulder, CO 80305, USA*
[b] *Statistical Engineering Division, National Institute of Standards and Technology, Boulder, CO, USA*

Abstract

We investigate the sources of uncertainty in the measurement of the reflection high-energy electron diffraction (RHEED) intensity oscillations during growth of AlAs, GaAs, and AlGaAs on GaAs substrates, and the resulting effects on predicted growth rates and composition. Sources of error examined include beam positioning, flux transients, substrate size, 'beat' phenomena in the RHEED oscillations, substrate temperature, and incident beam direction. We find that flux transients and flux nonuniformity are the dominant systematic errors in predicting growth rates and composition with RHEED. From flux uniformity measurements, we estimate the beam positioning error for our growth system to be 0.2–0.6%/mm, and substrate size to impact the uncertainty by as much as several percent. In addition to these errors, flux transients can cause an uncertainty of up to 1%. We also present a procedure that uses the measured variance in the growth rates to calculate the composition with the smallest mean square error.

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1. Introduction

The use of reflection high-energy electron diffraction (RHEED) intensity oscillations has proven to be a powerful tool to understand growth mechanisms of GaAs, AlAs and AlGaAs in molecular beam epitaxy (MBE). Early studies [1–3] of RHEED patterns generated discussion of the surface constructions and growth mechanisms of the GaAs system, but one important feature, immediately recognized by all, is the close relationship between the RHEED intensity oscillations and the growth rate of their films. Specifically, the period of the intensity oscillations of the specular diffraction spot corresponds to the time required to grow exactly 1 ML of crystal over a broad range of growth conditions. Great effort has gone into understanding the RHEED intensity oscillations [4,5], and their usefulness in monitoring MBE growth. Nevertheless, there is considerable variability in how the RHEED technique is applied. The purpose of this paper is to critically examine

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*Corresponding author. Tel.: +303-497-3340; fax: +303-497-3387.

E-mail address: harvey@boulder.nist.gov (T.E. Harvey).

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the sources of uncertainty in MBE measurements of growth rate and composition using RHEED intensity oscillations and to isolate which practices will minimize systematic and random error.

This study is organized in two main parts, namely exploration of the systematic effects on the measurements and the statistical propagation of uncertainties. The systematic effects examined include beam positioning, substrate size, substrate temperature and incident beam direction. We find that flux transients and flux nonuniformity are the dominant systematic errors in predicting growth rates and composition with RHEED. Included in the flux nonuniformity is uncertainty due to the 'beat' phenomenon sometimes observed in RHEED oscillations. These errors are minimized primarily by using a small substrate, accurate beam positioning, and careful analysis of the data. The statistical uncertainty section includes both data on typical noise and uncertainty in RHEED oscillation growth rate measurements and a discussion of which methods will give the best estimate of film composition.

2. Experimental procedure

The MBE system in this experiment has a turbo molecular pump, load lock, and buffer chamber for wafer degassing. The chamber has nine source locations, with standard Knudsen cells for Ga and Al, and a valved As effusion cell with a high-temperature cracker. The substrate is radiatively heated, and the substrate temperature is monitored using a commercial optical pyrometer. For this set of experiments, the substrate temperature was 580–600°C, and the V/III beam equivalent pressure ratio (measured using an ion filament beam flux monitor) was maintained at approximately 18:1. The system is designed to accommodate single wafers up to 76 mm diameter. For our measurements of RHEED oscillation intensity, we used two square GaAs specimens with 20 and 10 mm edge lengths, mechanically held at wafer center with molybdenum plates. The system has a standard RHEED gun mounted 28 cm from wafer center and produces a 7.0 keV electron beam with grazing incident angle of approximately 2°. The phosphor screen is mounted ~26 cm from wafer center. We have a video camera to record the RHEED pattern on the phosphor screen and a small CRT monitor to display the pattern. We use a small photodiode mounted on the monitor to measure the oscillations of the RHEED specular spot. The intensity signal from the photodiode is amplified, filtered at 3 Hz, and sampled at 60 Hz for computer acquisition. Unless otherwise stated, the azimuth angle of the substrate was 5–10° off the [0 1 1] direction.

We archive most of the RHEED measurements that we make for our growth runs, giving us a large pool of data for statistical analysis. The data are plotted and analyzed using computer algorithms to locate the maxima and minima of the intensity oscillations. The instantaneous oscillation period is derived from two successive maxima or minima. We make 3–7 measurements for the Ga, Al, and Al + Ga (both shutters open) fluxes, each measurement containing 50–200 oscillations. Each AlGaAs layer is covered immediately with GaAs, and the surfaces are allowed to recover to a uniform, smooth condition between successive flux measurements.

3. Results and discussion

Because the RHEED oscillation data is acquired while the specimen is stationary, the results are particularly susceptible to variations in growth rate across the specimen. These variations are of substantially less consequence during the growth run, when the substrate is rotating. Although larger RHEED specimens better simulate the actual wafer in thermal environment and produce more reliable signals for optical pyrometry, the larger beam path, in this case, is subject to greater inaccuracies arising from spatially varying growth rates. The most obvious inaccuracy is that the growth rate at the substrate center is the one that most closely approximates the growth rate that will be produced on a larger rotating substrate. This means that the electron beam must be located in the center of the RHEED substrate. By moving the electron beam, we measured a growth rate variation of $0.3 \pm 0.2\%/\mathrm{mm}$ for the Ga and

$0.5 \pm 0.4\%$/mm for the Al in a direction perpendicular to the electron beam path. Thickness measurements of periodic structures characterized with X-ray diffraction and optical reflectivity show spatial variations that are within a factor of two of these values. These values are, of course, specific to the machine and evaporation cell geometry. We note that the layers grown in the same machine with substrate rotation display a thickness variation of less than 0.05%/mm and a compositional uniformity better than the limits of detection (<0.001 in Al mole fraction x) in the central $20\,\text{mm} \times 20\,\text{mm}$ region of the substrate.

Variation of the growth rate along the electron beam also influences the observed growth rate. The primary manifestation is interference between the signals from different spatial regions of the sample whose rates of oscillation gradually become out of phase with each other as growth progresses. The interference leads to damping of the oscillations and, in some cases, 'beats' appear in which the oscillation amplitude recovers after becoming very small [6]. An observed beat in the RHEED oscillation curve from a GaAs specimen $20\,\text{mm}^2$ is illustrated in Fig. 1. The variation of the spot intensity can be modeled in a simple way by integrating cosine contributions from each small spatial region along the electron beam. The RHEED intensity for a linear variation $\pm \mathrm{d}G$ in growth rate G is predicted as $I(t) = \cos(2\pi Gt)[\sin(2\pi\,\mathrm{d}Gt)/(2\pi\,\mathrm{d}Gt)]$. This model ignores any decay in intensity from increased diffuse scattering and also weighs all the spatial regions evenly. Nevertheless, as shown in Fig. 1, the model does predict damping of the intensity amplitude and beats after about 25 oscillations, even for spatial variations on the order of a few percent. Applied to a square specimen with an edge length of 20 mm, the $\pm 1.9\%$ growth rate variations used in the model correspond to 0.2%/mm.

Fig. 1 illustrates another important consideration of accuracy when the data is taken on large specimens. The model shows that $I(t)$ goes through a phase change relative to the main carrier wave, $\cos(2\pi Gt)$, at each beat, effectively losing one half of a cycle. A common procedure for analyzing RHEED oscillations is to select two strong peaks widely spaced in time, and then to divide the number of oscillations separating these peaks by their time separation to obtain the

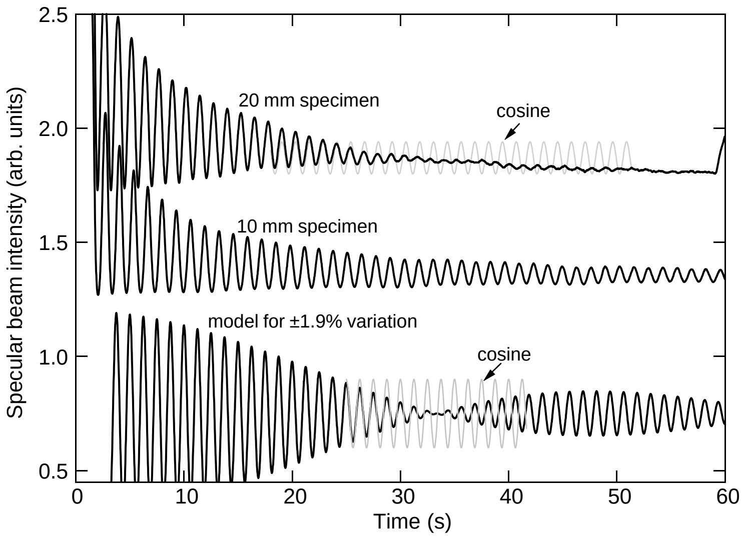

Fig. 1. Specular beam intensity as a function of time for specimens of two different sizes illustrating 'beats' for the larger specimen due to variations of growth rate along the beam path. Also shown are results of a modeling $\pm 1.9\%$ growth rate variation (see text for model details). The cosine function overlay on the beat model illustrates how the intensity curve switches phase when going through a beat, effectively losing one half-cycle.

growth rate in monolayers (ML) per second. The loss of one half-cycle would introduce an error if peaks on either side of the beat are selected and the number of oscillations between them is assumed to be an integer. For a half-cycle error every 25 cycles, similar to the data shown in Fig. 1, the resulting growth rate error is 2%. Finally from Fig. 1, we note that the 10 mm sample shows oscillations that decay in time much more slowly than for the 20 mm sample. This longevity is particularly important in evaluating flux transients.

Fig. 2 shows another potential source of uncertainty in predicting growth rates from the RHEED oscillations, namely the phenomenon of flux transients. These changes in instantaneous growth rate occur when the cell cools or heats because of changes in its thermal environment, particularly when a shutter is first opened to initiate growth. In Fig. 2, we observe a peak in the Al growth rate roughly 10 s after the shutter is opened. The growth rates for the different time periods shown in Fig. 2 vary by up to 1.5% for the Al cell. Data acquired by averaging over shorter time periods will display even larger variations. Thus, ignoring the effects of flux transients will lead to errors in determination of the long-term growth rate by up to 3%. The best prediction for growth rate in subsequent runs depends on the time needed to grow the layers in those runs. For layers taking several minutes or more to grow, the average growth rate measured at times long after the shutter opening would be the most accurate measure. For very thin layers such as those of quantum wells and dots, much care needs to be taken in order to accurately predict actual thickness and composition.

Continuing the study of systematic effects in the RHEED intensity oscillations, we also examined the effect of substrate orientation on the oscillation period. We measure the intensity oscillations with the RHEED electron beam path several degrees off the [0 1 1] direction, then repeated the measurement several degrees off the [0 $\underline{1}$ 1] direction, for the Ga, Al, and Al + Ga fluxes. The data show no statistically significant difference in average oscillation period between the two orientations. We also find, in agreement with other reports [5], that there is no statistically significant difference between the sum of the Al and Ga

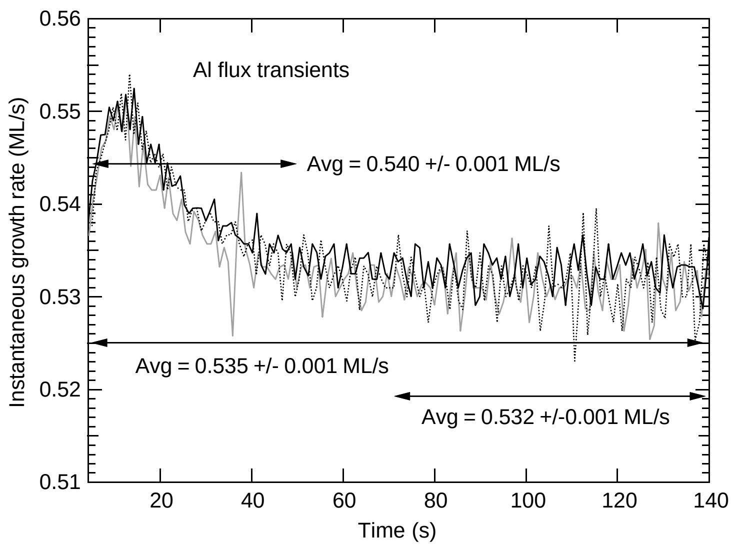

Fig. 2. Instantaneous growth rate as a function of time for successive AlAs RHEED oscillation curves. The average of the instantaneous growth rates are displayed on the graph next to the line showing the time interval over which the averages were calculated.

oscillations measured separately and the Al + Ga oscillations. Finally, we measured all three growth rates at substrate temperatures ranging from 595°C to 622°C. These growth rates are identical within the measurement of the uncertainty range, indicating that there is not significant re-evaporation of either Al or Ga from the surface at these temperature ranges. Based on these results, we conclude that RHEED growth rate measurements are most accurate when the RHEED specimen is small (10 mm or less) and when the electron beam is placed within 1 mm of the center of the substrate mount. Conditions must also be chosen so that flux transient effects can be properly compensated. A summary of the errors associated with each effect is given in Table 1.

Composition of the film can also be extracted from the RHEED measurements of the AlAs, GaAs, and AlGaAs growth rates, a, g, and b, respectively. These three growth rates can be combined to calculate the Al mole fraction x in four different ways, which are: a/b, $(b-g)/b$, $a/(a+g)$, and $(b-g)/(a+g)$. The mean square error in the estimation of x depends on both any bias (systematic error) in the growth rate measurements and the noise in the growth rate measurements. We carried out statistical analysis of our data and of these equations to determine which equations will propagate the least mean square error. The noise in the data is assessed by taking successive RHEED oscillation curves, typically three or four of each type. For each curve, the average instantaneous growth rate at long times, hereafter called the average growth rate, is computed. The standard deviations of sets of average growth rates are plotted with hollow symbols in Fig. 3(a). The instantaneous growth rate curves are noisy at long times, and the standard deviations of the instantaneous growth rate distributions within single curves are also plotted in Fig. 3(a) with solid symbols. Data acquired when the RHEED screen is heavily coated with As is particularly likely to have a large amount of noise at long times. The reproducibility of the average growth rate over successive curves is nevertheless quite good, indicating that the standard deviations of the average growth rates represent a reasonable measure of the maximum fluctuations in the actual growth rate.

Using Taylor-series expansions, the standard deviation in Al mole fraction x is estimated in

Table 1
Contribution of various factors to uncertainty in RHEED growth rate measurements for specimens of two sizes

Factor	Uncertainty (%)				Comments
	10 mm		20 mm		
Overall uncertainty	1	4	4	7	Includes 0.5% short-time reproducibility standard deviation and any factors with uncertainty at least this large
Electron beam position, in center half of specimen		3		6	Depends on cell flux spatial distribution
Electron beam within 1 mm of center	0.5		0.5		Depends on cell flux spatial distribution
Interference beats	<0.4		2		20 mm value assumes data spans beat
Flux transients, uncontrolled		2		3	Varies up or down by factor of 2 depending on cell conditions. Generally larger for Al
Flux transients measured	<0.4		3		Cannot reach stable cell conditions before oscillations die out for 20 mm specimen
Temperature	<0.3		<0.3		595–622°C
Reconstruction (2× versus 4×)	<0.4		<0.4		Assumes beam on same spot after substrate rotation

Overall uncertainty is calculated by adding other significant sources in quadrature. Substrate temperature and surface reconstruction direction used during RHEED acquisition are not distinguishable from the short-time reproducibility uncertainty.

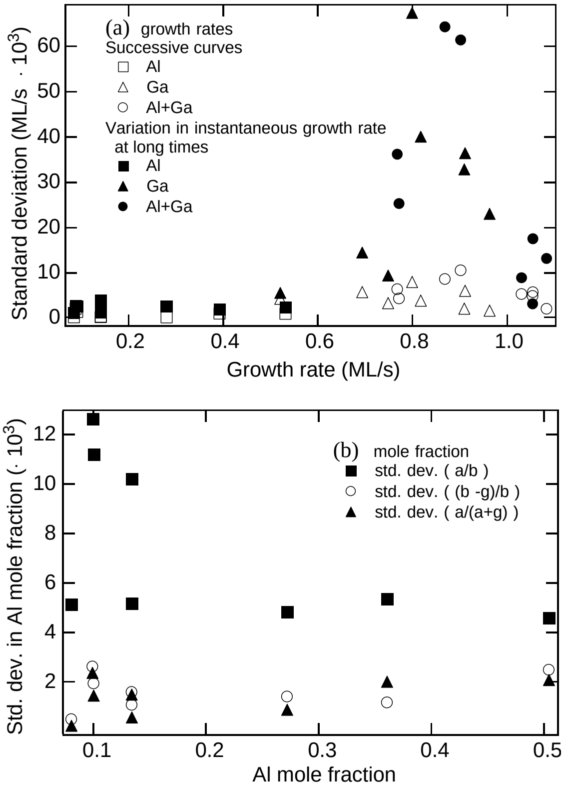

Fig. 3. (a) Standard deviation for RHEED growth rate data. Hollow symbols are the standard deviation of the average growth rates determined from a set of three or four consecutive oscillation curves. Solid symbols are the standard deviation of the instantaneous growth rate values within a single curve at long times. (b) Standard deviation of the aluminum mole fraction x for the data sets in (a) for three different methods of calculating mole fraction from the growth rate measurements. For most data sets, the equation $x = a/(a+g)$ gives the best estimate of the mole fraction.

terms of the mean and variance of the growth rates a, g, and b, for each of the four equations listed above. The equation $(b-g)/(a+g)$ gives higher mean square error than at least one of the other equations regardless of the values of the standard deviations of the average growth rates. The standard deviations σ for x based on the other equations can be estimated as follows:

$$\sigma(a/b) \approx (a/b)\,\mathrm{sqrt}\,\{(\sigma_a/a)^2 + (\sigma_b/b)^2\},$$

$$\sigma((b-g)/b) \approx (a/b)\,\mathrm{sqrt}\,\{(\sigma_b^2+\sigma_g)^2/a^2 + (\sigma_b/b)^2 - 2\sigma_b^2/ab\},$$

$$\sigma(a/(a+g)) \approx (a/b)\,\mathrm{sqrt}\,\{(\sigma_a/a)^2 + (\sigma_a^2+\sigma_g^2)/b^2 - 2\sigma_a^2/ab\}. \quad (1)$$

These standard deviations in x have been calculated from the standard deviations in average growth rate (hollow symbols in Fig. 3(a)), and the results are given in Fig. 3(b). The equation $x = a/(a+g)$ propagates the least error for most of our data, although the equation $x = (b-g)/b$ gives similar results. The error estimate in the mole fraction for both of these equations is typically $\sigma = 0.002$.

4. Conclusions

We have estimated the magnitude of errors in the measurements associated with RHEED intensity oscillations. With care, the overall uncertainty in growth rate can be reduced to the level of 1–2%. The best results are obtained with the smallest substrate possible, which reduces the sensitivity of the technique to spatial variations of the effusion cell fluxes. The smaller size also facilitates placing the beam as close to the center of the sample as possible, where the growth rate is most representative of the growth rate that will be obtained with substrate rotation. The error associated with spatial deviation from the center is approximately 0.5%/mm. We have shown typical data sets with flux transient effects and discuss the possible increase in the uncertainty of up to 3%. Finally, through an analysis of variance we show that the equation $a/(a+g)$ propagates the least error in the prediction of the composition of the AlGaAs layer for typical standard deviations in the average growth rates.

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